Anomalous Periodicity of the Current-Phase Relationship of Grain-Boundary Josephson Junctions in High-T_c Superconductors

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The current-phase relation (CPR) for asymmetric 45° Josephson junctions between two d-wave superconductors has been predicted to exhibit an anomalous periodicity. We have used the single-junction interferometer to investigate the CPR for this kind of junctions in $YBa_2Cu_3O_{7-x}$ thin films. Half-fluxon periodicity has been experimentally found, providing a novel source of evidence for the d-wave symmetry of the pairing state of the cuprates.

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There is growing evidence in favor of the $d_{x^2-y^2}$-wave symmetry of the pairing state of the high-temperature superconductors.[1] An unconventional pairing state requires the existence of zeros of the order parameter in certain directions in momentum space. Thermodynamic and spectroscopic measurements do indeed suggest their existence, but by themselves they do not exclude conventional s-wave pairing with nodes.[1] Direct evidence for the d-wave pairing state is provided by phase-sensitive experiments, which are based on the Josephson effect.[2] Quite generally, the current-phase relationship (CPR) of a Josephson junction, $I(\varphi)$, is an odd periodic function of φ with a period 2π.[3] Therefore $I(\varphi)$ can be expanded in a Fourier series

$$I(\varphi) = I_1 \sin\varphi + I_2 \sin 2\varphi + \ldots . \tag{1}$$

In the tunnel limit we can restrict ourselves to the first two terms in Eq. (1). Since the order parameter is bound to the crystal lattice, $I(\varphi)$ of a weak link depends on the orientation of the d-wave electrodes with respect to their boundary. The existing phase-sensitive experiments exploit possible sign changes of I_1 between different geometries.[2] In this Letter we present a new phase-sensitive experimental test of the symmetry of the pairing state of the cuprates. Namely, in certain geometries, the I_1 term should vanish by symmetry. In such cases, the CPR should exhibit an anomalous periodicity.

Let us analyze the angular dependence of $I_{1,2}$ in a junction between two macroscopically tetragonal d-wave superconductors. As emphasized in Ref. 4, also heavily twinned orthorhombic materials such as $YBa_2Cu_3O_{7-x}$ belong to this class, if the twin boundaries have odd symmetry. We consider first an ideally flat interface between the superconducting electrodes. Let θ_1 (θ_2) denote the angle between the normal to the grain boundary and the a axis in the electrode 1 (2), see Fig. 1. If we keep only the lowest-order angular harmonics, the symmetry of the problem dictates that[4]

$$I_1 = I_c \cos 2\theta_1 \cos 2\theta_2 + I_s \sin 2\theta_1 \sin 2\theta_2. \tag{2}$$

The coefficients I_c, I_s are functions of the barrier strength, temperature T, etc. The I_2 term results from higher-order tunneling processes and we neglect its weak angular dependence. It is seen from Eq. (2) that the criterion for the observation of an anomalous CPR, $I_1 = 0$, is realized for an asymmetric 45° junction, i.e. a junction with $\theta_1 = 45°$ and $\theta_2 = 0$. For an interface which is not ideally flat, $\theta_i = \theta_i(x)$ are functions of the coordinate x along the junction. $I_1 = 0$ remains valid even in this case, if the average values $\langle\theta_1(x)\rangle = 45°$ and $\langle\theta_2(x)\rangle = 0$.

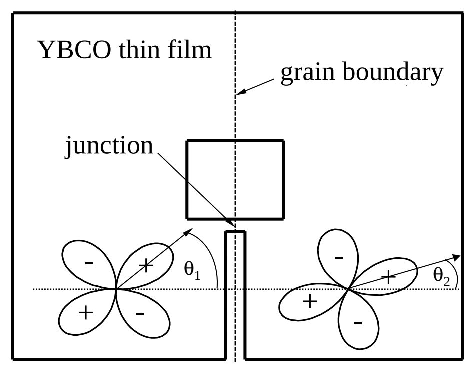

FIG. 1. Sketch of the interferometer (not in scale).

The I_2 term is present also in weak links based on conventional s-wave superconductors but for all known types of weak links $|I_2/I_1| < 1$. For instance, for a tunnel junction $|I_2/I_1| \ll 1$. For a SNS junction, $I \propto \sin\varphi/2$ at $T = 0$,[5] and the Fourier expansion Eq. (1) leads to $I_2/I_1 = -2/5$. Therefore, a possible experimental observation of $|I_2/I_1| \gg 1$ in an asymmetric 45° junction provides direct evidence of d-wave symmetry of the pairing state in the cuprates.

We have investigated the CPR of $YBa_2Cu_3O_{7-x}$ thin film bicrystals with asymmetric 45° [001]-tilt grain boundaries[6–8] as sketched in Fig. 1, using a single-junction interferometer configuration in which the

Josephson junction is inserted into a superconducting loop with a small inductance L. In a stationary state without fluctuations, the phase difference φ across the junction is controlled by applying external magnetic flux Φ_e penetrating the loop: $\varphi = \varphi_e - \beta f(\varphi)$. Here $\varphi_e = 2\pi\Phi_e/\Phi_0$ is the external flux normalized to the flux quantum $\Phi_0 = 2.07 \times 10^{-15}$ Tm^2. The CPR is written as $I(\varphi) = I_0 f(\varphi)$, where I_0 is the maximal Josephson current. $\beta = 2\pi L I_0/\Phi_0$ is the normalized critical current. In order to obtain the CPR for the complete phase range $-\pi \leq \varphi \leq \pi$ the condition $\beta < 1$ has to be fulfilled, because for $\beta > 1$ the curve $\varphi(\varphi_e)$ becomes multivalued and there are jumps of φ and a hysteresis for a sweep of φ_e. Following Ref. 3, we express the effective inductance of the interferometer using the derivative f' with respect to φ as $L_{int} = L[1+1/f'(\varphi)]$. The inductance can be probed by coupling the interferometer to a tank circuit with inductance L_T, quality factor Q, and resonance frequency ω_0.[9] External flux in the interferometer is produced by a current $I_{dc} + I_{rf}$ in the tank coil and can be expressed as $\varphi_e = 2\pi(I_{dc}+I_{rf})M/\Phi_0 = \varphi_{dc}+\varphi_{rf}$, where $M^2 = k^2 L L_T$, and k is a coupling coefficient. Taking into account the quasiparticle current in the presence of voltage V across the junction the phase difference is given by the relation $\varphi = \varphi_{dc} + \varphi_{rf} - \beta f(\varphi) - 2\pi\tau(\varphi)V/\Phi_0$, where $\tau(\varphi) = L/R_J(\varphi)$ and $R_J(\varphi)$ is the resistance of the junction . In the small-signal limit $\varphi_{rf} \ll 1$ and in the adiabatic case $\omega\tau \ll 1$, keeping only the first-order terms, the effective inductance L_{eff} of the tank curcuit-interferometer system reads

$$L_{eff} = L_T\left(1 - k^2\frac{L}{L_{int}}\right) = L_T\left(1 - \frac{k^2\beta f'(\varphi)}{1+\beta f'(\varphi)}\right).$$

Thus the phase angle α between the driving current and the tank voltage U at the resonant frequency of the tank circuit ω_0 is

$$\tan\alpha(\varphi) = \frac{k^2 Q\beta f'(\varphi)}{1+\beta f'(\varphi)}. \tag{3}$$

Using the relation $[1+\beta f'(\varphi)]d\varphi = d\varphi_{dc}$ valid for $\varphi_{rf} \ll 1$ and $\omega\tau \ll 1$, one can find the CPR from Eq. (3) by numerical integration.

The advantage of the measurement of the CPR of an asymmetric 45° junction with respect to the by-now standard phase-sensitive tests of pairing symmetry based on the angular dependence of I_1 is twofold. First, it avoids the complications of the analysis of experiments caused by the presence of the term I_s.[4] Second, a flux trapped in the SQUID does not invalidate the conclusions about the ratio $|I_2/I_1|$ and hence about the pairing symmetry, while this is not the case in standard phase-sensitive tests of the d-wave symmetry of the pairing state.[10]

The films of thickness 100 nm were fabricated using standard pulsed laser deposition on (001) oriented $SrTiO_3$ bicrystalline substrates with asymmetric [001] tilt misorientation angles $45° \pm 1°$. They were subsequently patterned by Ar ion-beam etching into 4×4 mm^2 square washer single-junction interferometer structures (Fig. 1). The widths of the junctions were $1 \div 2$ μm. The washer square holes had a side-length of 50 μm. This geometry of the interferometer gives $L \approx 80$ pH. The resistance of the junction is higher than 1 Ω and the condition for the adiabatic limit $\omega\tau \ll 1$ is satisfied. For measurements of $\alpha(\varphi_{dc})$, several tank circuits with inductances $0.2 \div 0.8$ μH and resonance frequencies $16 \div 35$ MHz have been used. The unloaded quality factor of the tank circuits $70 < Q < 150$ has been measured at various temperatures. The coupling factor k was determined from the period ΔI_{dc} of $\alpha(I_{dc})$ using $M\Delta I_{dc} = \Phi_0$. Its value varied between 0.03 and 0.09. The amplitude of I_{rf} was set to produce the flux in the interferometer lower than $0.1\Phi_0$.

The measurements have been performed in a gas-flow cryostat with a five-layer magnetic shielding in the temperature range $4.2 \leq T < 90$ K. The experimental setup was calibrated by measuring interferometers of the same size with 24° and 36° grain boundaries. We have studied 5 samples, out of which sample No. 1 exhibited the most anomalous behavior. Samples Nos. 2,3 were less anomalous and the remaining two samples had high critical currents and their $I(\varphi)$ was conventional. In Fig. 2 we plot the phase angle α as a function of the dc current I_{dc} for samples Nos. 1,2. The data for the 36° junction is shown for comparison. Note that at $T = 40$ K the periodicity of $\alpha(I_{dc})$ is the same for all samples.

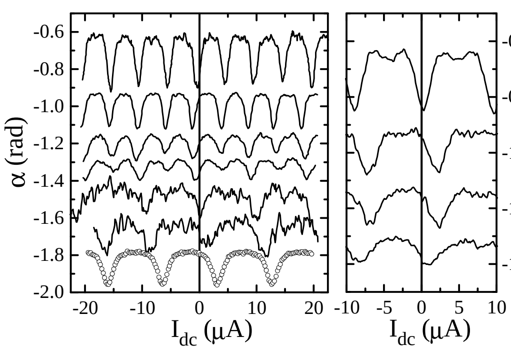

FIG. 2. Left panel: Phase angle between the driving current and the output voltage measured for the sample No. 1 at different temperatures as a function of the dc current I_{dc}. The curves are shifted along the y axis and the data for $T = 30$ and 40 K are multiplied by factor 4 for clarity. From top to bottom, the data correspond to T =4.2, 10, 15, 20, 30 and 40 K. The data measured on 36° bicrystals ($\theta_1 \approx 36°, \theta_2 \approx 0$) at T =40 K in the same washer geometry are shown for comparison (open circles). Right panel: The same for the sample No. 2. From top to bottom, the data correspond to T =4.2, 10, 15 and 20 K.

We assume that the period of $\alpha(I_{dc})$ at T =40 K, ΔI_{dc} =9.6 μA, corresponds to $\Delta\varphi_{dc} = 2\pi$. In order to determine the CPR we take $\varphi_{dc} = 0$ at a maximum or minimum of α. This is necessary in order to satisfy

$I(\varphi = 0) = 0$, as required by general principles.[3] The experimentally observed shift of the first local extreme of $\alpha(I_{dc})$ from $I_{dc} = 0$ (Fig. 2) can be due to flux trapped in the interferometer washer. Most probably, this flux resides in the long junction of the interferometer. The long junction does not play an active role because the Josephson penetration depth is much shorter than its length, and external fields producing I_{dc} are smaller than its critical field. Nevertheless the long junction sets the phase difference for $I_{dc} = 0$ at the small junction.

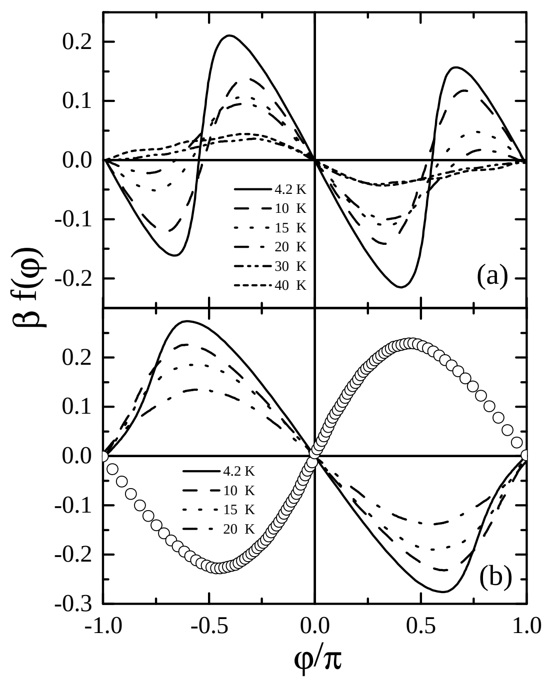

FIG. 3. a) Josephson current through the junction for the sample No. 1 as a function of the phase difference φ, determined from the data in Fig. 2. The statistics of $\alpha(\varphi)$ was improved by folding the data back to the interval $\langle 0, \pi\rangle$ and taking an average. The symmetry $\alpha(\varphi) = \alpha(-\varphi)$ was assumed. b) The same for the sample No. 2. The data for the asymmetric 36° bicrystal at $T = 40$ K (open circles) is also shown.

In Fig. 3, we show the CPR determined from the data in Fig. 2. For all curves we have performed a minimal necessary shift consistent with $I(\varphi = 0) = 0$. Thus, for the samples Nos. 1,2 we have assumed that at $\varphi_{dc} = 0$ a minimum of $\alpha(\varphi_{dc})$ is realized. For an interferometer with a conventional s-wave weak link (and also for the 36° junction), at $\varphi_{dc} = 0$ a maximum of $\alpha(\varphi_{dc})$ is realized. Note that the maximum (minimum) of $\alpha(\varphi_{dc})$ at $\varphi_{dc} = 0$ implies a diamagnetic (paramagnetic) response of the interferometer in the limit of small applied fields. In Fig. 4 we plot the coefficients I_1 and I_2 determined by Fourier analysis of the CPR for the sample No. 1 at various temperatures. With decreasing T, $|I_2|$ grows monotonically down to $T = 4.2$ K, while the I_1 component exhibits only a weak temperature dependence.

Our experimental results can be understood as follows. Deviations from ideal geometry of the asymmetric 45° junction, $\langle\theta_1\rangle = 45° + \alpha_1$ and $\langle\theta_2\rangle = \alpha_2$, lead to a finite value of I_1. Thus, imperfections of the junction increase its critical current. For this reason we believe that samples Nos. 2-5 contain imperfections and from now we concentrate on nearly ideal junctions (such as sample No. 1) with $|\alpha_1|, |\alpha_2| \ll 1$. For such junctions, the ratio I_2/I_1 exhibits the following temperature dependence. For $T \to 0$, $|I_2/I_1| \gg 1$. The region $T \sim T_c$ can be analyzed quite generally within Ginzburg-Landau theory. Let the electrodes be described by (macroscopic) order parameters $\Delta_{1,2} = |\Delta| e^{i\varphi_{1,2}}$. Then the phase-dependent part of the energy of the junction is $E = a[\Delta_1\Delta_2^\star + \text{H.C.}] + b[(\Delta_1\Delta_2^\star)^2 + \text{H.C.}] + \ldots$ where $a, b, \ldots$ depend weakly on T.[11] Thus for T close to T_c we estimate $I_1 \propto |\Delta|^2 \propto (T_c - T)$ and $I_2 \propto |\Delta|^4 \propto (T_c - T)^2$, leading to $|I_2/I_1| \ll 1$. These expectations are qualitatively consistent with the experimental data shown in Fig. 4.

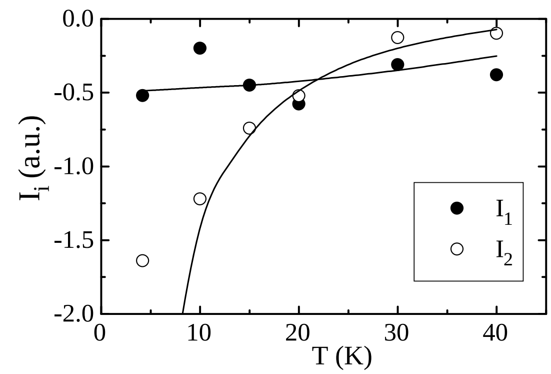

FIG. 4. Temperature dependence of the Fourier expansion coefficients $I_{1,2}$ determined from the experimental data in Fig. 3a. Solid lines are the Fourier expansion coefficients for the numerical data in Fig. 5.

So far, our discussion was based solely on symmetry arguments. Let us attempt a more quantitative analysis of our data now. Two different microscopic pictures of asymmetric 45° Josephson junctions between d-wave superconductors have been considered in the literature. The first picture assumes a microscopically tetragonal material and an ideally flat interface.[11–13] Within this picture, there are two contributions to the Josephson current. The first is due to bulk states and in the tunnel limit it is well described by the Sigrist-Rice term I_c in Eq. (2).[14] The second is due to mid-gap states which develop close to the surfaces of unconventional superconductors.[15] $I(\varphi)$ for the sample No. 1 calculated according to the model of Ref. 12 is shown in Fig. 5. The experimental data can be fitted by a relatively broad range of barrier heights. However, if we require the 36° junction to be fitted by the same (or smaller) barrier height as for the 45° junction, we conclude the barrier must be rather low.[16] The T dependence of $I(\varphi)$ requires

a choice of $T_c \approx 60$ K in the non-selfconsistent theory of Ref. 12. The reduction from the bulk $T_c = 90$ K is probably due to a combined effect of surface degradation and order-parameter suppression at the sample surface. The temperature dependence of the ratio of the π and 2π periodic components in $I(\varphi)$ is seen to be in qualitative agreement with experimental data in Fig. 3a. This is explicitly demonstrated in Fig. 4 where we compare the experimentally obtained $I_{1,2}$ with the results of the Fourier analysis of the curves in Fig. 5. The divergence of I_2 as $T \to 0$ is an artifact of the ideal junction geometry assumed in Ref. 12. If the finite roughness of the interface is taken into account, this divergence is cut off and the experimental data in Fig. 4 do indeed resemble theoretical predictions for a rough interface.[13] However, the nonselfconsistent theory of Ref. 12 is unable to explain the experimentally observed steep CPR close to the minima of the junction energy (see Fig. 3a). In the limit of vanishing barrier height, the theoretical CPR does have steep portions, but these are located close to the maxima of the junction energy (see also Ref. 11).

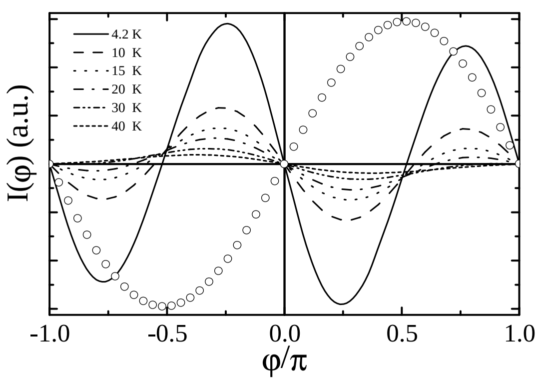

FIG. 5. $I(\varphi)$ calculated according to Eq. (64) of Ref. 12 for a junction with $\theta_1 = 45.5°$, $\theta_2 = 0$, $\lambda d = 1.5$, $\kappa = 0.5$, and $T_c = 60$ K. $I(\varphi)$ at $T = 40$ K for the 36° bicrystal (open circles) was calculated for the same parameters except for $\theta_1 = 36°$.

In a different approach to the asymmetric 45° junction, one assumes a heavily twinned orthorhombic material (which is macroscopically tetragonal, however) and/or a meandering interface with $\theta_i = \theta_i(x)$.[17,18] Hence the critical current density $j_c(x)$ is a random function with a typical amplitude $\langle |j_c(x)| \rangle \sim j_c$. If the average critical current along the junction $\langle j_c \rangle \ll j_c$, a spontaneous flux is generated in the junction, and $|I_2/I_1| \gg 1$.[17,18] In particular, for $\langle \theta_1 \rangle = 45°$ and $\langle \theta_2 \rangle = 0$, there is an equal amount of parts having positive and negative j_c, leading to $\langle j_c \rangle = 0$ and $I_1 = 0$. Note that also within the picture of Refs. 17,18, the d-wave symmetry of the pairing state is crucial, otherwise the condition $\langle j_c \rangle \ll j_c$ is difficult to satisfy.

Our present understanding of $I(\varphi)$ in the asymmetric 45° junction is only qualitative. There is considerable experimental evidence[6–8] that the grain boundary junctions are at most piecewise flat. However, we cannot say whether the shape of $I(\varphi)$ is dominated by the mid-gap states in the microscopically flat regions, or by spontaneous flux generation due to the spatial inhomogeneity of the junction. This issue requires further study.

In conclusion, we have measured the magnetic field response of a single-junction interferometer based on asymmetric 45° grain-boundary junctions in $YBa_2Cu_3O_{7-x}$ thin films. Half-fluxon periodicity has been experimentally found, in agreement with theoretical predictions for $d_{x^2-y^2}$-wave superconductors. Hence, our results provide a novel source of evidence for the d-wave symmetry of the pairing state in the cuprates.

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